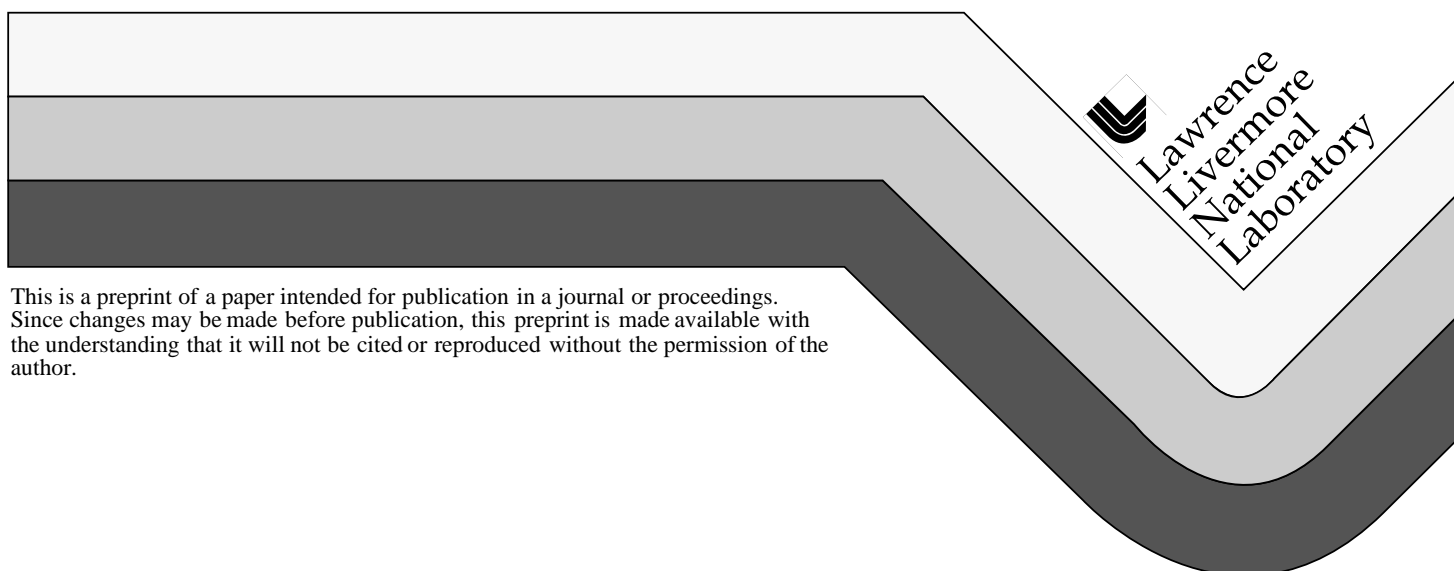


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Reversible (Unitized) PEM Fuel Cell Devices

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Abstract

Regenerative fuel cells (RFCs) are enabling for many weight-critical portable applications, since the packaged specific energy (>400 Wh/kg) of properly designed lightweight RFC systems is several-fold higher than that of the lightest weight rechargeable batteries. RFC systems can be rapidly refueled (like primary fuel cells), or can be electrically recharged (like secondary batteries) if a refueling infrastructure is not conveniently available. Higher energy capacity systems with higher performance, reduced weight, and freedom from fueling infrastructure are the features that RFCs promise for portable applications. Reversible proton exchange membrane (PEM) fuel cells, also known as unitized regenerative fuel cells (URFCs), or reversible regenerative fuel cells, are RFC systems which use reversible PEM cells, where each cell is capable of operating both as a fuel cell and as an electrolyzer. URFCs further economize portable device weight, volume, and complexity by combining the functions of fuel cells and electrolyzers in the same hardware, generally without any system performance or efficiency reduction. URFCs are being made in many forms, some of which are already small enough to be portable. Lawrence Livermore National Laboratory (LLNL) has worked with industrial partners to design, develop, and demonstrate high performance and high cycle life URFC systems. LLNL is also working with industrial partners to develop breakthroughs in lightweight pressure vessels that are necessary for URFC systems to achieve the specific energy advantages over rechargeable batteries. Proton Energy Systems, Inc. (Proton) is concurrently developing and commercializing URFC systems (UNIGENTM product line), in addition to PEM electrolyzer systems (HOGENTM product line), and primary PEM fuel cell systems. LLNL is constructing demonstration URFC units in order to persuade potential sponsors, often in their own conference rooms, that advanced applications based on URFCs are feasible. Safety and logistics force these URFC demonstration units to be small, transportable, and easily set up, hence they already prove the viability of URFC systems for portable applications.

Introduction

Energy storage systems with extremely high specific energy (>400 Wh/kg) have been designed that use lightweight pressure vessels to contain the gases generated by PEM-based reversible (unitized) regenerative fuel cells (URFCs). URFC systems are being designed and developed for a variety of applications (Figure 1), including high altitude long endurance (HALE) solar rechargeable aircraft (SRA), zero emission vehicles (ZEVs), hybrid energy storage/propulsion systems for spacecraft, energy storage for remote (off-grid) power sources, peak shaving for on-grid applications, and portable power systems (1-15). Portable RFC systems offer all of the features of portable fuel cells with the additional feature of being independent of a refueling infrastructure. The ability to electrically recharge the fuel cell powered device by water electrolysis drastically simplifies operations, eliminating the need for fuel supply logistics. In URFC systems, the electrolysis is performed using the same PEM cells that produce power, resulting in systems with reduced weight, volume, cost, and complexity.

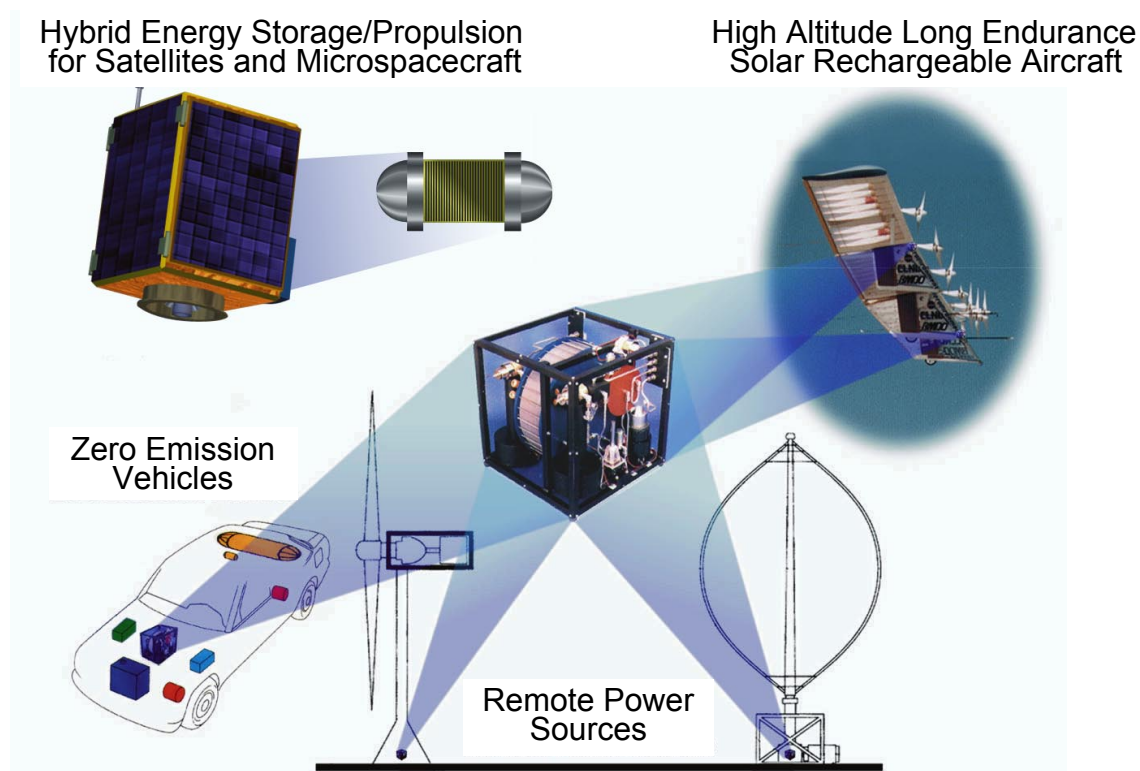


Figure 1. URFC Systems Have Mobile and Stationary Applications

Reversing the electrochemical reaction that converts hydrogen and oxygen to water without changing hardware is a tougher development problem than unidirectional power generation, but one that is already solved. The appropriate proprietary catalysts have been demonstrated to have high cycle life. No system performance or efficiency is lost compared to either fuel cells or electrolyzers constructed in this reversible geometry. The substitution of a URFC for a

conventional fuel cell favors hydrogen as a fuel, stored as compressed gas in lightweight (high performance) cylinders. URFC systems which are typically recharged electrically (as opposed to physically refueled) may favor the storage/consumption of pure oxygen, rather than air, in order to maximize performance and minimize maintenance. However, weight, volume, efficiency, complexity, safety, operational environment, and maintenance costs all need to be carefully considered before choosing between an oxygen system and an air-breathing system. Due to the additional sophistication required to develop RFC systems, few manufacturers have developed them to date. Of these, Proton Energy Systems, Inc. is currently the only vendor of commercial URFC systems (Figures 3 and 4), while United Technologies Corporation, Hamilton Standard Space and Sea Systems Division, serves primarily aerospace applications, as described in another paper in these proceedings (1). Other small US companies, such as Giner, Inc. (16) and Lynntech Inc. (17) may be capable of supplying suitably-catalyzed PEM electrode assemblies for URFCs.

The convenience of electrical recharging opens many of the current applications for secondary batteries to fuel cell powered devices. The addition of solar panels to supply that electricity frees the device entirely from the logistics of refueling. This approach has been taken by LLNL for advanced aircraft and spacecraft (Figure 1). The option to have freedom from a refueling infrastructure is enabling for applications like emergency power, recreation (e.g., camping, recreational vehicles, boating, etc.), field operations (forestry, mining, surveying, non-studio photography, etc.), and almost any portable application that must ride in a vehicle. Various fuels being considered for fuel cell systems (even methanol) may be difficult to obtain in remote locations, and a widespread hydrogen infrastructure will not be available in the near-term. Clean water and electricity are much simpler to obtain anywhere, and closed system energy storage applications don't even need to obtain water, since their reactants are conserved.

Early History of PEM Devices

Electrolyzers and fuel cells that utilize proton exchange membrane (PEM) technology have undergone continued development for rigorous aerospace and military applications since the late 1950's (18). These applications required that systems operate with unquestioned long-term reliability in environments that often imposed severe mechanical challenges due to shock, vibration, or high fluid pressures (19-21).

Products that utilize PEM technology enjoy a rich aerospace and commercial heritage. PEM technology was invented at the General Electric Company (GE) in the late 1950's and matured at both GE and Hamilton Standard over the next several decades, resulting in reliable, high technology product solutions for rigorous challenges of many military and aerospace applications (1).

The first of these was NASA's Gemini space mission in which PEM fuel cells manufactured at GE provided power to the space capsule for astronaut life support and instrument operation. Components for these systems were hand-made by highly skilled aerospace technicians working in a laboratory environment. This mode of fabrication translated to a high cost structure associated with manufacture of PEM fuel cells at that time.

PEM fuel cell applications that followed the Gemini mission included power systems for the Biosatellite mission, high altitude aerostats, and sonobuoys, just to name a few. Although these were important applications, the need for PEM fuel cells declined in the late 1960's and into the 1970's resulting in a drop in the number of people engaged in developing PEM fuel cells from greater than 500 to less than 100. GE began examining alternative markets for this enabling technology and discovered that a number of applications for PEM electrolysis existed and could prove fruitful from an overall business standpoint.

In the early to mid-1970's GE began working in earnest to develop PEM water electrolysis technology for undersea life support. This led to the development of the U.S. Navy Oxygen Generating Plant, a state-of-the-art electrolyzer that produces 225 standard cubic feet per hour (6370 standard liter per hour) of oxygen at 3000 psi. The British Royal Navy quickly adopted this technology in the early 1980's and deployed PEM electrolysis systems to outfit its entire submarine fleet. The service record has been excellent, with well over 1 million cell hours accumulated on electrolysis stacks operating in service at sea without a single cell failure. Success in the development and manufacture of electrolyzers for military life support led GE to initiate the development of a small benchtop-sized commercial hydrogen generator for laboratory chromatography applications. This product, now manufactured by Packard Instrument Company, has been highly successful in competing against liquid caustic electrolyte systems with over 10,000 commercial units delivered to date.

Proton Commercializes PEM Products

Since the mid-1980's PEM fuel cell development has branched into commercial applications including transportation and stationary power. The steep cost reduction imperative of these commercial applications now requires engineers to design fuel cell hardware for minimum cost while maintaining system performance and reliability. Low-cost PEM water electrolyzers, such as the Proton Energy Systems Inc. HOGEN™ series (Figure 2), are currently being commercialized for industrial hydrogen gas generation applications. A growing number of important industrial processes such as heat treating, silicon wafer production, and argon purification can now receive pure, high pressure hydrogen "by wire" via PEM water electrolysis at a cost lower than hydrogen delivered conventionally by tank truck.

HOGEN™ 300

HOGEN™ 10



Figure 2. Proton's HOGEN™ 10 and 300 Hydrogen Generators are Being Delivered

Proton Energy Systems, Inc. (Proton) was founded in 1996 for the sole purpose of commercializing PEM products for gas generation and energy storage applications. Proton's first family of products, the HOGEN™ series of water electrolyzers, economically produces very pure hydrogen and oxygen gas from water (22). Gases analyzed from Proton's HOGEN™ 300 unit have shown hydrogen purities well in excess of 99.9995%. Proton's HOGEN™ family of hydrogen generators now includes units having 10 and 300 standard cubic feet per hour (280 and 8500 standard liter per hour) hydrogen production capacities. These fully weatherized units are completely automated and therefore require minimal operator interaction. Facility interfaces are minimized with only power and potable water being required. HOGEN™ hydrogen generator units can be used to produce hydrogen for metals heat treating, microelectronics manufacture, argon purification, turbine cooling, renewable energy production, food processing, and a host of other applications. Units of this size are especially cost competitive with conventional hydrogen supply due to the high fixed cost of hydrogen truck transport and on-site customer storage.

LLNL Develops Advanced URFC Applications

Significant developments in PEM fuel cells and electrolyzers have been combined over the past two years to form the basis for revolutionary, efficient, energy storage systems. The breakthrough demonstrations of such energy storage systems have been funded by the U.S. DOE and U.S. utilities to provide electricity on demand from renewable energy resources (e.g., wind and solar

power). These energy storage systems replace secondary batteries, and in some cases hydroelectric reservoirs, to produce high quality power when the primary energy source (e.g., sunlight or wind power) is unavailable (7).

Table 1. URFCs offer higher packaged specific energy than secondary batteries.

Storage System	Theoretical Specific Energy [Wh/kg]	Packaged Specific Energy [Wh/kg]	Comments
H₂/O₂ URFC	3660	400-1000	URFC with lightweight pressure vessels
Li-SPE/MO_x	735	220	Li-solid polymer electrolyte/ metal oxide, novel packaging
Ag/Zn	450	200	Excess Zn required for high cycle life, low charge rate
Li/LiCoO₂	735	150	Poor cycle life, high capacity fade
Li/AlFeS₂	515	150	≥400°C thermal management
Na/S	1180	150	~350°C thermal management
Li/TiS₂	470	130	~50% DOD for high cycle life (900 cycles)
Li/ion	700	100 (135 ^a)	Projection revised Nov. 1996
Ni/Zn	305	90	Excess Zn required, low specific energy
Ni/MH_x	470	70 (85 ^a)	MH _x is metal hydride, projection revised Nov. 1996
Ni/H₂	470	60	Low specific energy
Ni/Cd	240	60	Low specific energy
Pb/acid	170	50	Low specific energy

^a Projection revised Nov. 1996, private communication, B.M. Barnett (A.D. Little, Inc.)

Aerospace applications can already afford to use PEM systems, and LLNL did not hesitate to conclude that URFCs exchanging gases with lightweight pressure vessels would make a superior energy storage system for solar powered aircraft. LLNL found that URFC-based energy storage would be advantageous in other vehicular and utility applications (some of which are illustrated in Figure 1). Tank performance factor (burst pressure x internal volume / tank weight = $P_b V/W$) improvements have provided increased system specific energy over the already singularly high values for URFC systems (10,23). This is particularly important for weight-critical systems for aerospace, portable applications, and the huge automotive market. One space application is likely to fund development of PEM stacks in the 100-200 Watt range that would be suitable for many portable

applications. Although LLNL has designed RFC systems using URFCs and discrete PEM fuel cells and electrolyzers, the reduced mass and complexity of URFCs often proves superior or enabling in energy storage applications.

These electrochemical energy storage systems are functionally equivalent to secondary batteries yet retain the highly attractive features of fuel cells, including high specific energy and long lifetimes. Unlike batteries, URFCs uncouple both the oxygen electrode and the hydrogen electrode from direct participation in the storage chemistry. These electrodes support the reactions but are not transformed by them, thereby permitting external storage of reactants. This separation of energy storage and energy conversion frees URFC-based energy storage from size, weight, and cost penalties of batteries.

URFC-based energy storage allows energy storage densities approaching a significant fraction of the theoretical energy storage limits of hydrogen (Table 1) which, by weight, is the highest potential chemical energy storage medium. The hydrogen that embodies chemical energy storage in these systems must be contained, and therefore only with lightweight hydrogen storage concepts can the packaged energy storage density achieve significant advances over rechargeable batteries. The URFC can also electrolyze and store gases at high pressure, without mechanical compression. Such high pressure storage of hydrogen in lightweight pressure vessels will allow these URFC-based systems to meet the needs of volume-constrained applications.

Proton Develops Commercial URFC Applications

The inadequacies of secondary battery systems in effectively meeting energy storage requirements for many commercial applications prompted Proton to initiate the development of URFCs. Proton has developed liquid feed URFC energy storage for Electric Power Research Institute (EPRI) and static feed URFC energy storage systems for NASA Glenn Research Center (Figure 3).

Under a program funded by EPRI, Proton Energy Systems, Inc. has begun development of URFCs for utility, premium power, and remote-site applications where reliable, efficient, low cost energy conversion and storage is necessary. This system, known as the UNIGEN™, finds application in off-grid renewable energy storage, on-grid load leveling/peak shaving and zero-emissions transportation systems. Its smaller off-grid applications overlap portable applications because of the difficulties of bringing in and setting up RFC energy storage systems in remote locations. The compact and rugged cell design employed in UNIGEN™ systems (Figure 4) has performance characteristics in both modes that are comparable to dedicated fuel cells and electrolyzers.

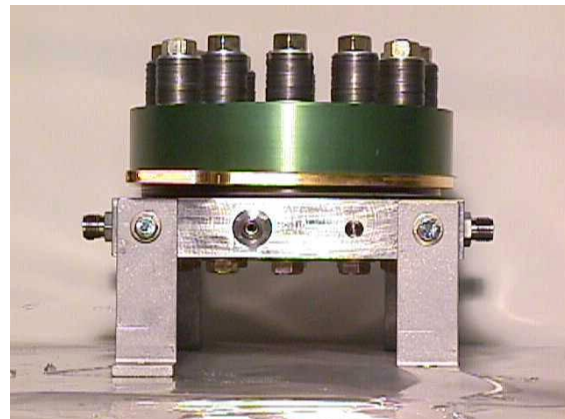


Liquid Feed URFC System for EPRI

Static Feed URFC for NASA

Figure 3. Proton Has Built Liquid Feed and Static Feed URFC (UNIGEN™) Systems

The UNIGEN™ is a particularly enabling technology for many critical energy storage applications. UNIGEN™ systems can be integrated with wind, solar, or hydroelectric power sources to provide clean, reliable power when the renewable power source is unavailable. This arrangement can provide an electrical infrastructure where none currently exists. As long as such a power source is small enough to be carried, it is ideal for portable applications. The UNIGEN™ can also provide 'premium' power for critical applications such as telecommunications in the event of an outage or fluctuation in the main power supply. Other applications include power augmentation in situations where day/night price differentials exist, and true zero emissions transportation systems.

URFC 2-cell stack (46 cm², 0.69 MPa)URFC 1-cell stack (93 cm², 2.8 MPa)**Figure 4. Examples of Proton's Liquid Feed URFC (UNIGEN™) Cell Stacks**

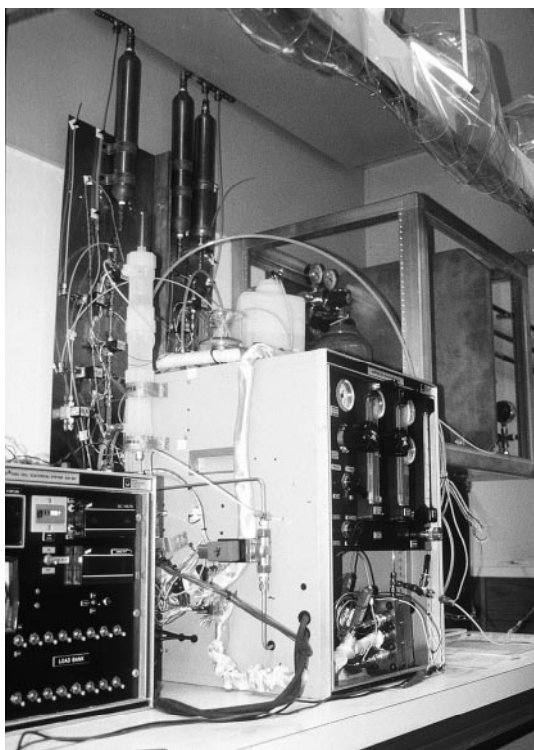
URFC Advantages

A URFC is just a reversible type of a proton exchange membrane (PEM) cell that functions without compromise as an advanced fuel cell, but is also designed so that *the same components* can operate in reverse to perform electrolysis of water. URFCs achieve high reliability and high performance using a PEM as the sole electrolyte. In the power generation (discharge) mode, H_2 and O_2 react to release energy and form by-product water. This water is retained in the system and is electrolyzed during the charge mode to generate H_2 and O_2 .

Current Proton cells can now generate H_2 and O_2 gases at pressures suitable for gaseous storage up to 400 psi without compressors. Generation at higher pressures (up to 6000 psi) has been demonstrated by others (1). Oxygen can be either stored as pressurized gas or supplied from ambient air. When operated under identical conditions, systems that use pressurized oxygen show a significantly higher efficiency at a given performance level (or a significantly higher performance at a given efficiency), than those that consume air. Safety aspects of high pressure oxygen need to be carefully assessed for individual applications. Proton's ultimate product is a complete URFC with integrated storage of the reactants through gas tankage, metal hydrides, or other emerging hydrogen storage media. LLNL cannot manufacture products, but can deliver (to its U.S. government sponsors) advanced energy storage systems based on the best available URFCs (including Proton's) and highest performance tankage.

LLNL URFC Demonstrations

URFC electrochemical research at LLNL commenced in 1996. A primary fuel cell test rig with a single cell (46 cm² active area) was modified and operated reversibly as a URFC for thousands of cycles (Figure 5). This URFC uses bifunctional electrodes (oxidation and reduction electrodes reverse roles when switching from charge to discharge, as with a rechargeable battery) for cathode feed electrolysis (water is fed from the hydrogen side of the cell) or anode feed electrolysis (water is fed from the oxygen side of the cell). The results of the cycle test demonstrated high cycle life with negligible degradation (3,8). Improved URFC cells, with high performance and reduced catalyst loading were demonstrated at LLNL, as shown in Figure 6 (3). Rapid cycle operation of URFC cells has been demonstrated at LLNL. Although cell internal hardware was designed in the early-1980's, fluid flow capabilities, control, electrical power instrumentation, and membrane electrode assemblies were added over several generations of upgrades. This URFC test facility is capable of cycling URFC cells using flow through reactants in fuel cell mode (hydrogen and oxygen or air), closed cycle reactants supplied by electrolysis products or high pressure gas storage cylinders, cathode feed electrolysis, or anode feed electrolysis.



LLNL's URFC Test Rig

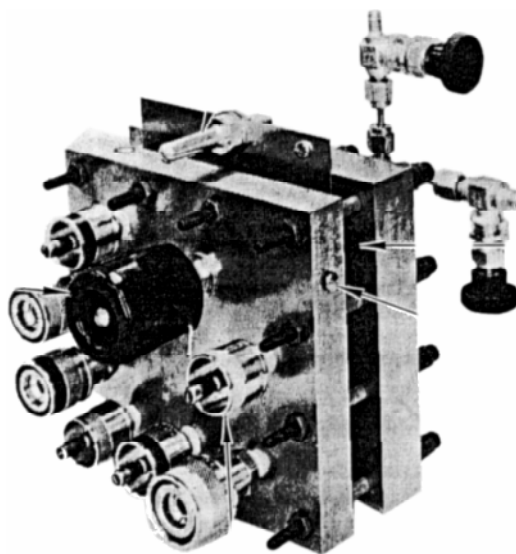
URFC Cell Stack (46 cm², 1.1 MPa)

Figure 5. URFC Test Rig and URFC Cell Stack at LLNL

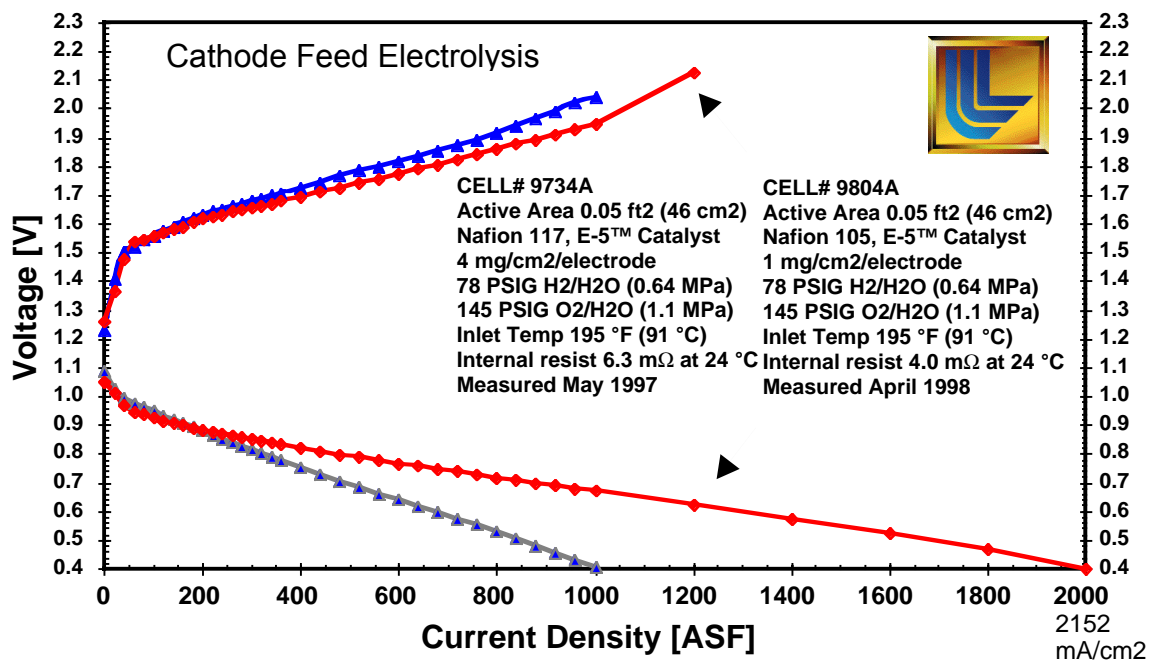


Figure 6. High Performance URFC Demonstrated With Reduced Catalyst Loading

Although the entire LLNL test rig can provide technical information of sufficient quality for publication, its complexity exceeds what would make sense for a portable energy storage system. Programmatic ‘sales’ often require taking the message that URFC technology is real into the conference rooms of sponsors. Neither speaker nor audience are willing to wait for hardware that takes more than ~15 minutes to operate. LLNL worked with Hamilton Standard to fabricate the first portable URFC demonstration unit in 1993, in order to help programmatic sales for the HALE SRA program (Figure 1). This LLNL program was funded by the Ballistic Missile Defense Organization (BMDO). Several copies of this URFC “propeller toy” were built which demonstrate that from an initially inert state (just low pressure water), electrolysis can commence (using solar power) to pressurize H₂ and O₂ storage tanks (all embodied in transparent polycarbonate). After a charging (pressurization) period of seconds to minutes, a switch is toggled to place the load (motor) across the fuel cell, so that it is powered by the URFC operating as a fuel cell. This URFC demonstration has proved to be so popular that its copies are invited to attend meetings and are sought by politicians, funding agents, and teachers worldwide. Proton recognized the sales value of such URFC ‘toys’ early, and has produced its own ‘toys’ for sales purposes (Figure 7) which may be useful in portable applications below ~5 Watts.



Figure 7. LLNL/Hamilton Standard URFC Demo (Left), Proton URFC Demo (Right)

After the HALE solar powered aircraft passed to NASA Dryden Research Center. However, both tankage and URFC technologies found support from spacecraft applications, many of which are small enough for their hardware to be easily portable. The URFC hydrogen and oxygen is the same ideal rocket propellant combination that is used by launch vehicles, such as the main engines of the Space Shuttle. The benign nature of water as a precursor for made-on-board rocket fuel has created the need for another generation of LLNL demonstrators (Figure 8). This “Water Rocket” demonstration or “torch toy” electrolyzes gases from initially ambient water up to 100 psi (0.69 MPa) H₂ and O₂. Once pressurized, the system is capable of demonstrating safe combustion in conference rooms, and has been designed to be combined with the URFC “propeller toy” to demonstrate hybrid energy storage and propulsion.

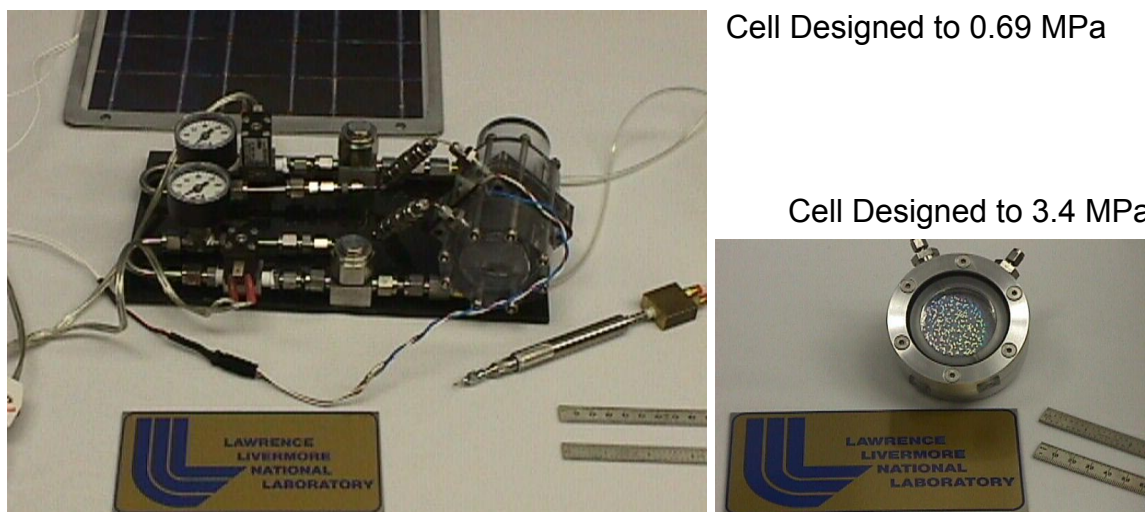


Figure 8. Conference Room Demonstrations of URFC Water Rocket Propulsion

The LLNL torch toy requires considerably more gas storage than the propeller toy to provide a long burn duration. Several seconds of quiet, almost invisible, millimeter-scale flame is hard for audiences to follow, but tens of seconds of burn duration provide the time to wave refractory metal wires through the flame to show its temperature, and provide a memorable display. This demonstrator is already portable, and has successfully met the requirements imposed by fielding even a small high temperature torch with pressurized hydrogen and oxygen. The ability to travel safely, and generate hydrogen and oxygen on demand from sunlight or slide projectors has already shown the URFC's potential infrastructure independence at perhaps a dozen meetings. A third generation of demonstration 'toys' (Figure 8) is being developed at LLNL with higher pressure capability, to illustrate URFC systems' independence of power and energy, power source and sink flexibility, and routine water refueling of bipropellant gas generators.

Summary

URFC technology is enabling for many portable applications because, unlike secondary batteries, the URFC permits the uncoupling of the power and energy aspects of the storage system. Thus, electrochemical components are sized to meet peak power demands while reactant storage is sized to meet total energy requirements for a given application. This rapidly advancing technology has a strong heritage based on over 35 years of development in PEM fuel cells and water electrolyzers, and is now ready to enable applications previously restricted by the performance of secondary batteries. Breakthroughs in lightweight tankage when coupled to lightweight URFC systems offer packaged specific energies that are several-fold higher than for the best secondary batteries. These systems can be rapidly refueled when the infrastructure is available, or can be electrically recharged, like a secondary battery, when freedom from refueling is necessary.

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